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SCIENCE

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X-RAYS AND CRYSTALLINE STRUCTURE¹

Two years have gone by since Dr. Laue made his surprising discovery of the interference effects accompanying the passage of X-rays through crystals. The pioneer experiment has opened the way for many others, and a very large amount of work, theoretical and practical, has now been done. As the preliminary exploration of the new country has proceeded, our first estimate of its resources has grown continuously; we have learned many things which help us to a better understanding of phenomena already familiar, and we have seen avenues of enquiry open out before us which as yet there has been little time to follow. The work is full of opportunities for exact quantitative measurements, where precision is sure to bring its due reward. There is enough work in sight to absorb the energies of many experimenters, and there is sure to be far more than we can see. When we consider the wideness of the new field, the quality and quantity of the work to be done in it, and the importance of the issues, we are scarcely guilty of over-statement if we say that Laue's experiment has led to the development of a new science.

The experiment itself—to put it very briefly—constitutes a proof that X-rays consist of extremely short ether waves. In order to appreciate the value of this demonstration, we must bear in mind the present conditions of our knowledge of the laws of radiation in general. Let us consider very shortly how the whole matter stood when the new work was begun.

When X-rays were first discovered eighteen years ago it was soon pointed out that they might consist of electro-magnetic disturbance of the ether analogous to those supposed to

¹ Read before the weekly evening meeting of the Royal Institution of Great Britain, June 5, 1914.

constitute light. It was true that the new rays seemed to be incapable of reflection, refraction, diffraction and interference, which were familiar optical phenomena. But it was pointed out by Schuster² that these defects could be explained as natural consequences of an extremely small wave-length. The positive evidence consisted mainly in the knowledge that the impact of the electrons on the anti-cathode of the X-ray bulb ought to be the occasion of electro-magnetic waves of some sort, and in the discovery by Barkla that the X-rays could be polarized, which last is a property also of light.

As experimental evidence accumulated, a number of results were found which the electro-magnetic theory was unable to explain, at least in a direct and simple manner. They were mainly concerned with the transference of energy from place to place. In some way or other the swiftly moving electron of the X-ray bulb transfers its energy to the X-ray, and the X-ray in its turn communicates approximately the same quantity of energy to the electron which originates from matter lying in the track of the X-ray, and which is apparently the direct cause of all X-ray effects. Experiment seemed to indicate that X-ray energy traveled as a stream of separate entities or quanta, the energy of the quantum differing according to the quality of the X-ray. It looked at one time as if it might be the simplest plan to deny the identity in nature of X-rays and light, to describe the former as a corpuscular radiation and the latter as a wave motion. Otherwise, it seemed that the electro-magnetic hypothesis would be torn to pieces in the effort to hold all the facts together.

But it appeared on a close examination of light phenomena also, though in much less obvious fashion, that the very same effects occurred which in the case of X-rays were so difficult to explain from an orthodox point of view. In the end it became less difficult to deny the completeness of the orthodox theory than the identity in nature of light and X-rays. Modern work on the distribution of energy in the spectrum, and the dependence of specific

heat upon temperature, has also led independently to the same point of view. It has been urged with great force by Planck, Einstein and others that radiated energy is actually transferred in definite units or quanta, and not continuously; as if we had to conceive of atoms of energy as well as of atoms of matter. Let it be admitted at once that the quantum theory and the orthodox theory appear to stand in irreconcilable opposition. Each by itself correlates great series of facts; but they do not correlate the same series. In some way or other the greater theory must be found, of which each is a partial expression.

The new discovery does not solve our difficulty at once, but it does two very important things. In the first place, it shows that the X-rays and light are identical in nature; in fact, it removes every difference except in respect to wave-length. The question as to the exact place where the difficulty lies is decided for us; we are set the task of discovering how a continuous wave motion, in a continuous medium, can be reconciled with discontinuous transferences of radiation energy. Some solution there must be to this problem. The second important thing is that the new methods will surely help us on the way to find that solution. We can now examine the X-rays as critically as we have been able to study light, by means of the spectrometer. The wavelength of the X-ray has emerged as a measurable quantity. The complete range of electro-magnetic radiations now lies before us. At one end are the long waves of wireless telegraphy, in the middle are first the waves of the infra-red detected by their heating effects, then the light waves, and then the short waves of the ultra-violet. At the other end are the extremely short waves that belong to X-radiation. In the comparative study of the properties of radiation over this very wide range we must surely find the answer to the greatest question of modern physics.

So much for the general question. Let us now consider the procedure of the new investigations, and afterwards one or two applications to special lines of enquiry.

The experiment due to Laue and his collab-

² *Nature*, January 23, 1896.

orators Friedrich and Knipping has already been described in this lecture room and is now well-known. A fine pencil of X-rays passes through a thin crystal slip and impresses itself on a photographic plate. Round the central spot are found a large number of other spots, arranged in a symmetrical fashion, their arrangement clearly depending on the crystal structure. Laue had anticipated some such effect as the result of diffraction by the atoms of the crystal. His mathematical analysis is too complicated to be described now, and indeed it is not in any circumstances easy to handle. It will be better to pass on at once to a very simple method of apprehending the effect which was put forward soon after the publication of Laue's first results. I must run the risk of seeming to be partial if I point out the importance of this advance, which was made by my son W. Lawrence Bragg. All the recent investigations of X-ray spectra and the examination of crystal structure and of molecular motions which have been carried out since then have been rendered possible by the easy grasp of the subject which resulted from the simpler conception.

Let us imagine that a succession of waves constituting X-radiation falls upon a plane containing atoms, and that each atom is the cause of a secondary wavelet. In a well known manner, the secondary wavelets link themselves together and form a reflected wave. Just so a sound wave may be reflected by a row of palings, and very short sound waves by the fibers of a sheet of muslin.

Suppose a second plane of atoms to lie behind the first and to be parallel to it. The primary wave weakened somewhat by passing through the first plane, is again partially reflected by the second. When the two reflected pencils join it will be of great importance whether they fit crest to crest and hollow to hollow, or whether they tend to destroy each other's effect. If more reflecting planes are supposed, the importance of a good fit becomes greater and greater. If the number is very large, then, as happens in many parallel cases in optics, the reflected waves practically annul each other unless the fit is perfect.

It is easily seen that the question of fit depends on how much distance a wave reflected at one plane loses in comparison with the wave which was reflected at the preceding plane: the fit will be perfect if the loss amounts to one, two, three, or more wave-lengths exactly. In its turn the distance lost depends on the spacing of the planes, that is to say, the distance from plane to plane, on the wavelength and on the angle at which the rays meet the set of planes.

The question is formally not a new one. Many years ago Lord Rayleigh discussed it in this room, illustrating his point by aid of a set of muslin sheets stretched on parallel frames. The short sound waves of a high pitched bird call were reflected from the set of frames and affected a sensitive flame; and he showed how the spacing of the planes must be carefully adjusted to the proper value in relation to the length of wave and the angle of incidence. Rayleigh used the illustration to explain the beautiful colors of chlorate of potash crystals. He ascribed them to the reflection of light by a series of parallel and regularly spaced twinning planes within the crystal, the distance between successive planes bearing roughly the same proportion to the length of the reflected wave of light as the distance between the muslin sheets to the length of the wave of sound.

Our present phenomenon is exactly the same thing on a minute scale: thousands of times smaller than in the case of light, and many millions of times smaller than in the case of sound.

By the kindness of Professor R. W. Wood I am able to show you some fine examples of the chlorate of potash crystals. If white light is allowed to fall upon one of them, the whole of it is not reflected. Only that part is reflected which has a definite wave-length or something very near to it, and the reflected ray is therefore highly colored. The wave-length is defined by the relation already referred to. If the angle of incidence is altered, the wave-length which can be reflected is altered, and so the color changes.

It is not difficult to see the analogy between

these cases and the reflection of X-rays by a crystal. Suppose for example that a pencil of homogeneous X-rays meets the cube face of such a crystal as rocksalt. The atoms of the crystal can be taken to be arranged in planes parallel to that face, and regularly spaced. If the rays meet the face at the proper angle, and only at the proper angle, there is a reflected pencil. It is to be remembered that the reflection is caused by the joint action of a series of planes, which, in this case, are parallel to the face; it is not a reflection by the face itself. The face need not even be cut truly: it may be unpolished or deliberately roughened. The reflection takes place in the body of the crystal, and the condition of the surface is of little account.

The allotment of the atoms to a series of planes parallel to the surface is not of course the only one possible. For example in the case of a cubic crystal, parallel planes containing all the atoms of the crystal may also be drawn perpendicular to a face diagonal of the cube, or to a cube diagonal, or in many other ways. We may cut the crystal so as to show a face parallel to any series, and then place the crystal so that reflection occurs, but the angle of incidence will be different in each case since the spacings are different. It is not necessary to cut the crystal except for convenience. If wave-length, spacing and angle between ray and plane are rightly adjusted to each other, reflection will take place in the crystal independently of any surface arrangement.

This is the "reflection" method of explaining the Laue photograph. W. L. Bragg showed in the first place that it was legitimate, and in the second that it was able to explain in the position of all the spots which Laue found upon his photographs. The different spots are reflections in different series of planes which may be drawn to contain the atoms of the crystal. The simpler conception led at once to a simpler procedure. It led to the construction of the X-ray spectrometer, which resembles an ordinary spectrometer in general form, except that the grating or prism is replaced by a crystal and the telescope by an

ionization chamber and an electroscope. In use a fine pencil of X-rays is directed upon the crystal, which is steadily turned until a reflection leaps out; and the angle of reflection is then measured. If we use different crystals or different faces of the same crystal, but keep the rays the same, we can compare the geometrical spacings of the various sets of planes. If we use the same crystal always, but vary the source of X-rays, we can analyze the latter, measuring the relative wave-lengths of the various constituents of the radiation.

We have thus acquired a double power: (1) We can compare the intervals of spacing of the atoms of a crystal or of different crystals, along various directions within the crystal; in this way we can arrive at the structure of the crystal. (2) We can analyze the radiation of an X-ray bulb; in fact we are in the same position as we should have been in respect to light if our only means of analyzing light had been by the use of colored glasses, and we had then been presented with a spectrometer, or some other means of measuring wave-length exactly.

We now come to a critical point. If we knew the exact spacings of the planes of some one crystal, we could now by comparison find the spacings of all other crystals, and measure the wave-length of all X-radiations; or if we knew the exact value of some one wave-length, we could find by comparison the values of all other wave-lengths, and determine the spacings of all crystals. But as yet we have no absolute value either of wave-length or of spacings.

The difficulty appears to have been overcome by W. L. Bragg's comparison of the reflecting effect in the case of rocksalt or sodium chloride, and sylvine or potassium chloride. These two crystals are known to be "isomorphous"; they must possess similar arrangements of atoms. Yet they display a striking difference both in the Laue photograph and on the spectrometer. The reflections from the various series of planes of the latter crystal show spacings consonant with an arrangement in the simplest cubical array, of which the smallest element is a cube at each corner of which is placed the same group, a single

atom or molecule, or group of atoms or molecules. In the case of rocksalt, the indications are that the crystal possesses a structure intermediate between the very simple arrangement just described and one in which the smallest element is a cube having a similar group of atoms or molecules at every corner and at the middle point of each face. The arrangement is called by crystallographers the face centered cube. The substitution of the sodium for the potassium atom must transform one arrangement into the other. This can be done in the following way, if we accept various indications that atoms of equal weight are to be treated as equivalent. Imagine an elementary cube of the crystal pattern to have an atom of chlorine at every corner and in the middle of each face, and an atom of sodium or potassium as the case may be, at the middle point of each edge and at the center of the cube. We have now an arrangement which fits the facts exactly. The weights of the potassium and chlorine atoms are so nearly the same as to be practically equivalent, and when they are considered to be so, the arrangement becomes the simple cube of sylvine. But when the lighter sodium replaces the potassium as in rocksalt the arrangement is on its way to be that of the face centered cube, and would actually become so were the weight of the sodium atoms negligible in comparison with those of chlorine. Of course the same result would follow were two or three, or any number of atoms of each sort to take the place of the single atom, provided the same increase were made in the number of the atoms of both sorts. We might even imagine two sorts of groups of chlorine and metal atoms, one containing a preponderance of the former, the other of the latter, but so that two groups, one of each kind, contain between them the same proportion of chlorine and metal as the crystal does. We must merely have two groups which differ in weight in the case of rocksalt, and are approximately equal in weight in the case of sylvine. But it was best to take the simplest supposition at the outset; and now the evidence that the right arrangement has been chosen is growing as fresh crystals are measured.

For it turns out that in all crystals so far investigated, the number of atoms at each point must always be the same. Why, then, should it be more than one? Or in other words, if atoms are always found in groups of a certain number, ought not that group to be called the atom?

As soon as the structure of a crystal has been found we can at once find by simple arithmetic the scale on which it is built. For we know from other sources the weight of individual atoms, and we know the total weight of the atoms in a cubic centimeter of the crystal. In this way we find that the nearest distance between two atoms in rocksalt is 2.81×10^{-8} cm., which distance is also the spacing of the planes parallel to a cube face. From a knowledge of this quantity the length of any X-ray wave can be calculated at once as soon as the angle of its reflection by the cube face has been measured. In other words, the spectrometer has now become a means of measuring the length of waves of any X-radiation, and the actual spacings of the atoms of any crystal.

From this point the work branches out in several directions. It will not be possible to give more than one or two illustrations of the progress along each branch.

Let us first take up the most interesting and important question of the "characteristic" X-rays. It is known that every substance when bombarded by electrons of sufficiently high velocity emits X-rays of a quality characteristic of the substance. The interest of this comparison lies in the fact that it displays the most fundamental properties of the atom. The rays which each atom emits are characteristic of its very innermost structure. The physical conditions of the atoms of a substance and their chemical associations are largely matters of the exterior: but the X-rays come from the interior of the atoms and give us information of an intimate kind. What we find is marked by all the simplicity we should expect to be associated with something so fundamental.

All the substances of atomic weight between about 30 and 120 give two strongly defined "lines"; that is to say, there are found among

the general heterogeneous radiation two intense almost homogeneous sets of waves. For instance, rhodium gives two pencils of wave-lengths, approximately equal to 0.61×10^{-8} cm. and 0.54×10^{-8} cm. respectively. More exactly the former of these is a close doublet having wave-lengths 0.619×10^{-8} and 0.614×10^{-8} . The wave-lengths of palladium are nearly 0.58×10^{-8} and 0.51×10^{-8} ; nickel 1.66×10^{-8} and 1.50×10^{-8} . Lately Moseley has made a comparative study of the spectra of the great majority of the known elements, and has shown that the two-line spectrum is characteristic of all the substances whose atomic weights range from that of aluminium, 27, to that of silver, 108. These X-rays constitute, there is no doubt whatever, the characteristic rays which Barkla long ago showed to be emitted by this series of substances.

Now comes a very interesting point. When Moseley sets the increasing atomic weights against the corresponding decreasing wave-lengths, the changes do not run exactly parallel with each other. But if the wave-lengths are compared with a series of natural numbers everything runs smoothly. In fact it is obvious that the steady decrease in the wave-length as we pass from atom to atom of the series in the periodic table implies that some fundamental element of atomic structure is altering by equal steps. There is excellent reason to believe that the change consists in successive additions of the unit electric charge to the nucleus of the atom. We are led to think of the magnitude of the nucleus of any element as being simply proportional to the number indicating the place of the element in the periodic table, hydrogen having a nuclear charge of one unit, helium two, and so on. The atomic weights of the successive elements do not increase in an orderly way; they mount by steps of about two, but not very regularly, and sometimes they seem absolutely to get into the wrong order. For example, nickel has an atomic weight of 58.7, whereas certain chemical properties and still more its behavior in experiments on radio-activity indicate that it should lie between cobalt (59) and copper (63.6). But the wave-lengths, which are now

our means of comparison, diminish with absolute steadiness in the order cobalt, nickel, copper. Plainly, the atomic number is a more fundamental index of quality than the atomic weight.

It is very interesting to find, in the series arranged in this way, four, and only four, gaps which remain to be filled by elements yet undiscovered.

Let us now glance at another and most important side of the recent work, the determination of crystalline structure. We have already referred to the case of the rocksalt series, but we may look at it a little more closely in order to show the procedure of crystal analysis.

The reflection of a pencil of homogeneous rays by a set of crystalline planes occurs, as already said, at a series of angles regularly increasing; giving as we say, spectra of the first, second, third orders, and so on. When the planes are all exactly alike and equally spaced the intensities of the spectra decrease rapidly as we proceed to higher orders, according to a law not yet fully explained. This is, for example, the case with the three most important sets of planes of sylvine, those perpendicular to the cube edge, the face diagonal, and the cube diagonal respectively. An examination of the arrangement of the atoms in the simple cubical array of sylvine shows that for all these sets the planes are evenly spaced and similar to each other. It is to be remembered that the potassium atom and the chlorine atom are so nearly equal in weight that they may be considered effectively equal. In the case of rocksalt the same may be said of the first two sets of planes, but not of the third. The planes perpendicular to the cube diagonal are all equally spaced, but they are not all of equal effect. They contain alternately, chlorine atoms (atomic weight 35.5) only and sodium atoms (atomic weight 23) only. The effect of this irregularity on the intensities of the spectra of different orders is to enhance the second, fourth, and so on in comparison with the first, third and fifth. The analogous effect in the case of light is given by a grating in which the lines are alternately light and

heavy. A grating specially ruled for us at the National Physical Laboratory shows this effect very well. This difference between rocksalt and sylvine and its explanation in this way constituted an important link in W. Lawrence Bragg's argument as to their structure.

When, therefore, we are observing the reflections in the different faces of a crystal in order to obtain data for the determination of its structure, we have more than the values of the angles of reflection to help us; we have also variations of the relative intensities of the spectra. In the case just described we have an example of the effect produced by want of similarity between the planes, which are, however, uniformly spaced.

In the diamond, on the other hand, we have an example of an effect due to a peculiar arrangement of planes which are otherwise similar. The diamond crystallizes in the form of a tetrahedron. When any of the four faces of such a figure is used to reflect X-rays, it is found that the second order spectrum is missing. The analogous optical effect can be obtained by ruling a grating so that, as compared with a regular grating of the usual kind, the first and second, fifth and sixth, ninth and tenth alone are drawn. To put it another way, two are drawn, two left out, two drawn, two left out, and so on. The National Physical Laboratory has ruled a special grating of this kind also for us, and the effect is obvious. The corresponding inference in the case of the diamond is that the planes parallel to any tetrahedral face are spaced in the same way as the lines of the grating. Every plane is three times as far from its neighbor on one side as from its neighbor on the other. There is only one way to arrange the carbon atoms of the crystal so that this may be true. Every atom is at the center of a regular tetrahedron composed of its four nearest neighbors, an arrangement best realized by the aid of a model. It is a beautifully simple and uniform arrangement, and it is no matter of surprise that the symmetry of the diamond is of so high an order. Perhaps we may see also, in the perfect symmetry and consequent effectiveness of the

forces which bind each atom to its place, an explanation of the hardness of the crystal.

Here, then, we have an example of the way in which peculiarities of spacing can be detected. There are other crystals in which want of uniformity both in the spacings and in the effective value of the planes combine to give cases still more complicated. Of these are iron pyrites, calcite, quartz and many others. It would take too long to explain in detail the method by which the structures of a large number of crystals have already been determined. Yet the work done already is only a fragment of the whole, and it will take no doubt many years, even though our methods improve as we go on, before the structures of the most complicated crystals are satisfactorily determined.

On this side then we see the beginning of a new crystallography which, though it draws freely on the knowledge of the old, yet builds on a firmer foundation since it concerns itself with the actual arrangement of the atoms rather than the outward form of the crystal itself. We can compare with the internal arrangements we have now discovered the external forms which crystals assume in growth, and the modes in which they tend to come apart under the action of solvents and other agents. By showing how atoms arrange and disarrange themselves under innumerable variations of circumstances we must gain knowledge of the nature and play of the forces that bind the atoms together.

There is yet a third direction in which enquiry may be made, though as yet we are only at the beginning of it. In the section just considered we have thought of the atoms as at rest. But they are actually in motion, and the position of an atom to which we have referred so frequently must be an average position about which it is in constant movement. Since the atoms are never exactly in their places, the precision of the joint action on which the reflection effect depends suffers materially. The effect is greater the higher the order of the spectrum. When the crystal under examination is contained within a suitable electric furnace and the atoms vibrate more violently

through the rise of temperature, the intensities of all orders diminish, but those of higher order much more than those of lower. The effect was foreseen by the Dutch physicist Debije, and the amount of it was actually calculated by him on certain assumptions. I have found experimental results in general accord with his formula. In passing it may be mentioned that as the crystal expands with rise of temperature the spacing between the planes increases and the angles of reflection diminish, an effect readily observed in practise.

This part of the work gives information respecting the movements of the atoms from their places, the preceding respecting their average positions. It is sure, like the other, to be of much assistance in the enquiry as to atomic and molecular forces, and as to the degree to which thermal energy is locked up in the atomic motions.

This brief sketch of the progress of the new science in certain directions is all that is possible in the short time of a single lecture: but it may serve to give some idea of its fascination and possibilities.

WILLIAM H. BRAGG

WALTER HOLBROOK GASKELL (1847-1914)

DR. WALTER HOLBROOK GASKELL, university lecturer on physiology and praelector on natural science at the University of Cambridge since 1883, died suddenly, after a short illness, on September 7, 1914. He came of a well-known Unitarian family in the north of England, and was born at Naples, on November 1, 1847. After receiving his preliminary education, he entered Trinity College, Cambridge, in 1865, subsequently taking a medical degree at University College, London, in 1878. At Cambridge, Gaskell was one of the earliest to come under the influence of Michael Foster, then praelector on physiology, and, at his instance, entered Ludwig's laboratory at Leipzig in 1874. Prior to Foster's advent, Gaskell had specialized in mathematics, being one of the wranglers in the Mathematical Tripos in 1869. From the date of his first paper, an important research on the vaso-di-

lator fibers of striated muscle,¹ the rest of his life was devoted to those researches on the motor mechanism of the heart and the sympathetic system which have made his name so well known in physiology and clinical medicine.

English physiology in the first half of the nineteenth century was represented mainly by the work of Sir Charles Bell (spinal nerve roots), Marshall Hall (reflex action), William Sharpey (ciliary motion), Sir William Bowman (theory of urinary secretion), William Prout (HCl in the gastric juice) and Thomas Graham (osmosis, colloids). In 1867 Michael Foster was Sharpey's assistant at University College, and, in 1870, at Huxley's instance, became praelector at Cambridge, while Burdon Sanderson became Waynflete professor of physiology at Oxford in 1882. From the teaching and inspiration of these two men came most of the brilliant names which have distinguished English physiology in the later period, with the exception of Starling, whose name is associated with Guy's Hospital. Schaefer was a Sharpey pupil, and was persuaded by Foster to devote his life to research. Leonard Hill and Gotch were Oxford men. From Cambridge came Langley, Henry Head, Sherrington, Roy, Adami, Gowland Hopkins and Gaskell.

When Gaskell began to work with Ludwig, every one believed in the so-called neurogenic theory of the heart's action, introduced by Borelli in 1680, viz., that the heart's movements, beat and tonus are due to nervous impulses. A little before Borelli, Harvey, in his demonstration of the circulation of the blood (1628), had advanced the idea that the heart is a muscular force pump, propelled by its own internal heat. This mystic dogma (the myogenic theory) was stated in more modern form by Haller in 1757, viz., that the heart's contraction is due to an inherent "irritability" of its muscle, the stimulus being the entrance and passage of venous blood through it. Both theories, neurogenic and myogenic, have had their ups and their downs to date. The neurogenic theory was resisted by Legallois in

¹ Proc. Roy. Soc. Lond. 1877, XXV., 439-445.